Measurement of the 36Cl deposition flux in central Japan: natural background levels and seasonal variability

Measurement of the ^{36}Cl deposition flux in central Japan: natural background levels and seasonal variability

Yuki Tosaki[a,b,*], Norio Tase[c], Kimikazu Sasa[b], Tsutomu Takahashi[b], Yasuo Nagashima[b]

[a]*Crustal Fluid Research Group, Geological Survey of Japan, National Institute of Advanced Industrial Science and Technology (AIST), Tsukuba, Ibaraki 305-8567, Japan*

[b]*AMS (Accelerator Mass Spectrometry) Group, Tandem Accelerator Complex, Research Facility Center for Science and Technology, University of Tsukuba, Tsukuba, Ibaraki 305-8577, Japan*

[c]*Faculty of Life and Environmental Sciences, University of Tsukuba, Tsukuba, Ibaraki 305-8572, Japan*

*Corresponding author. Tel./fax: +81 29 861 3087.

E-mail address: yuki.tosaki@aist.go.jp (Y. Tosaki)

Postal address: AIST Tsukuba Central 7, 1-1-1 Higashi, Tsukuba, Ibaraki 305-8567, Japan

Journal of Environmental Radioactivity, 106, 73–80.

Abstract

Essential parameters for the applications of ^{36}Cl as a tracer in groundwater studies include the initial ^{36}Cl/Cl ratio, at the time of recharge, and/or the natural background deposition flux of ^{36}Cl in the recharge area. To facilitate the hydrological use of ^{36}Cl in central Japan, this study aims to obtain a precise estimate of the long-term average local ^{36}Cl flux and to characterize its seasonal variability. ^{36}Cl in precipitation was continuously monitored in Tsukuba, central Japan

over a period of >5 years. The ^{36}Cl flux showed a clear seasonal variation with an annual peak during the spring, which is attributed to the seasonal variability of tropopause height. The long-term average ^{36}Cl flux (32 ± 2 atoms m^{-2} s^{-1}), estimated from the measured data, is consistent with the prediction from the ^{36}Cl latitudinal fallout model scaled using the global mean production rate of 20 atoms m^{-2} s^{-1}. The initial $^{36}Cl/Cl$ ratio was estimated to be $(41 \pm 6) \times 10^{-15}$, which is similar to that of pre-bomb groundwater in the Tsukuba Upland. An observation period covering an 11-year solar cycle would yield more accurate estimates of the values, given the increased ^{36}Cl flux during the solar minimum.

Keywords: ^{36}Cl, flux, precipitation, accelerator mass spectrometry

1. Introduction

The long-lived radionuclide ^{36}Cl, with a half-life of 301,000 years, has been successfully used as an environmental tracer in groundwater studies (e.g., Phillips and Castro, 2003). The widespread use of ^{36}Cl in such studies is largely attributed to the advent of accelerator mass spectrometry (AMS) in the late 1970s (Elmore et al., 1979). Because of the extremely low abundance of ^{36}Cl in the environment (typically 10^{-12}–10^{-15}), AMS is practically the only technique capable of detecting and quantifying natural levels.

Hydrological investigations using ^{36}Cl have been thoroughly reviewed in previous studies (Bentley et al., 1986a; Fabryka-Martin et al., 1987; Fontes, 1989; Fontes and Andrews, 1994; Phillips, 2000). Owing to the long half-life of ^{36}Cl, its most common application is the dating of very old groundwaters in large-scale aquifers such as the Great Artesian Basin of Australia (Bentley et al., 1986b; Torgersen et al., 1991; Love et al., 2000), the Milk River aquifer in Alberta, Canada (Phillips et al., 1986; Nolte et al., 1990, 1991), and the Aquia aquifer in

Maryland, USA (Purdy et al., 1987, 1996). The ^{36}Cl bomb pulse, which originates from nuclear tests in the 1950s, has also been used to trace young groundwater (Cook and Robinson, 2002; Tosaki et al., 2011b). Essential parameters for both of these applications include the initial $^{36}Cl/Cl$ ratio, at the time of recharge, and/or the natural background deposition flux of ^{36}Cl in the recharge area.

Atmospheric ^{36}Cl is predominantly produced in the stratosphere (~54%–90%; Huggle et al., 1996; Masarik and Beer, 1999) by cosmic-ray-induced spallation of ^{40}Ar at a global mean rate of approximately 20 atoms m^{-2} s^{-1} (Huggle et al., 1996; Masarik and Beer, 1999, 2009). The aerosol-bound ^{36}Cl and gaseous $H^{36}Cl$ in the stratosphere are transported out of the stratosphere and across the tropopause within ~2 years (Synal et al., 1990). They are removed quickly from the troposphere by wet or dry deposition, while mixing with stable chlorine in the atmosphere. The mean residence time of ^{36}Cl in the troposphere is expected to be on the order of weeks, according to estimates of atmospheric aerosol residence times (Turekian et al., 1977; Bleichrodt, 1978; Raisbeck et al., 1981).

Atmospheric chlorine is mainly of oceanic origin, and as a result decreases exponentially with increasing distance from the coast (Eriksson, 1960). Consequently, the $^{36}Cl/Cl$ ratios in precipitation and infiltrating water are controlled mainly by the latitudinal pattern of atmospheric ^{36}Cl deposition (Lal and Peters, 1967; Bentley et al., 1986a) and the delivery of marine-derived Cl. As an example, the distribution of $^{36}Cl/Cl$ ratios over the continental United States has been estimated by Bentley et al. (1986a), who found low values in coastal areas and higher values in the interior. This distribution pattern has been confirmed by measured $^{36}Cl/Cl$ values for pre-bomb groundwaters (Davis et al., 2003; Moysey et al., 2003). Once the initial $^{36}Cl/Cl$ value is known, the ^{36}Cl flux can be reasonably estimated by using a mass balance equation for ^{36}Cl and Cl at the ground surface (Andrews et al., 1994).

The spatial distribution of ^{36}Cl in precipitation during the spring of 2007 was investigated on a continental scale in Europe (Johnston and McDermott, 2008), yielding a distribution of the $^{36}Cl/Cl$ ratio similar to that in the United States (Moysey et al., 2003). Thus, the most direct approach in obtaining the initial value and/or the background flux is to monitor ^{36}Cl in precipitation (for several years to obtain reasonable estimates) (Davis et al., 1998). However, only a few studies have continuously measured ^{36}Cl deposition fluxes for 1–2 years, at a time resolution finer than monthly (Hainsworth et al., 1994; Knies et al., 1994; Santos et al., 2004). These earlier studies revealed a seasonal trend in the ^{36}Cl flux, with a peak in spring due to air exchange between the stratosphere and the troposphere. A longer observation period would allow seasonal changes to be distinguished from short-term fluctuations.

For evaluating ^{36}Cl flux, chlorine recycling as CH_3Cl is another issue that should be mentioned. Ice core data from Greenland (Synal et al., 1990) showed that the bomb-derived enhanced ^{36}Cl flux returned to pre-bomb level in around 1985. In a global-scale study (Scheffel et al., 1999; Blinov et al., 2000), however, measured ^{36}Cl flux around the early 1990s was still up to one order of magnitude higher than model prediction (Lal and Peters, 1967). They attributed it to storage of chlorine (including bomb-produced ^{36}Cl) in the biosphere and its subsequent release in the form of CH_3Cl into the troposphere. Later in Europe in 2007, Johnston and McDermott (2008) concluded that ^{36}Cl flux has returned to pre-bomb value.

This study aims to obtain a precise estimate of the long-term average local ^{36}Cl flux and to characterize its seasonal variability. The results will facilitate the use of ^{36}Cl in hydrological applications in central Japan. In addition, we investigate the possible influence of chlorine recycling as CH_3Cl, which may limit the use of ^{36}Cl as a young groundwater tracer. Our study, which covers a period of >5 years, highlights a characteristic seasonal variation in local ^{36}Cl flux, and constrains estimates of the annual average ^{36}Cl flux and the initial $^{36}Cl/Cl$ value.

2. Materials and methods

2.1. Sample collection

Monthly bulk precipitation samples were collected over a period of >5 years (from April 2004 to December 2009) on the roof of the Physical Sciences Building, University of Tsukuba (36°06′N, 140°06′E), located on the Tsukuba Upland in central Japan (Fig. 1). The climate of the area is humid temperate, with an annual precipitation of 1246 ± 178 mm and an annual mean temperature of 13.4 ± 0.6°C (average values for 1951–2000; meteorological data at the Aerological Observatory in Tsukuba, which is located 6 km southeast of the sampling site). The precipitation collector consists of a polyethylene (PE) funnel (diameter of 15 or 21 cm), a high-density polyethylene (HDPE) bottle (3 or 5 L), a specially designed piece of glassware with a ping-pong ball inside (to prevent evaporative loss of the sample; e.g., Shimada et al., 1994; Yabusaki et al., 2006), and silicon plugs used to make connections. Two collectors were usually set up to ensure sufficient monthly precipitation sample volumes for the measurement of ^{36}Cl, even in cases where the monthly precipitation amount was significantly smaller than normal.

2.2. Analytical methods

After collection, the sample volumes were measured and aliquots were extracted for the analysis of major dissolved ions. In cases where the sample volume of one bottle was not sufficient for the analysis of ^{36}Cl, two bottles of the sample were combined before the volume measurement. The samples for dissolved ion analysis were passed through 0.20 μm membrane filters (25HP020AN, Advantec, Tokyo, Japan). For the samples collected between April and November 2004, Cl^- concentrations were measured by ion chromatography (QIC Analyzer, Dionex, Sunnyvale, CA, USA) at the Watershed Environment Laboratory, Graduate School of

Life and Environmental Sciences, University of Tsukuba. The remaining samples (collected between December 2004 and December 2009) were analyzed for the concentrations of Na^+, Mg^{2+}, Ca^{2+}, and Cl^- by ion chromatography (Ion Analyzer IA-100, Dkk-Toa, Tokyo, Japan) at the Tandem Accelerator Complex, Research Facility Center for Science and Technology, University of Tsukuba.

Prior to ^{36}Cl analysis, samples were first filtered through glass microfiber filters (GF/B, Whatman, Maidstone, UK) and then through 0.45 μm membrane filters (JHWP04700, Millipore, Billerica, MA, USA). An ion exchange system was developed to concentrate sample chloride. The system includes a solution-sending pump (Masterflex L/S variable speed modular drive, No. 7553-80, Cole-Parmer, Vernon Hills, IL, USA) with four pump heads and four anion exchange columns. Each pump head uses Tygon tubing (inner diameter, 3.1 mm) to transport the sample into a column. Two systems enabled simultaneous processing of eight samples.

Approximately 20 mL (wet volume) of AG 4-X4 anion exchange resin in the free-base form (100–200 mesh; Bio-Rad Laboratories, Hercules, CA, USA) was loaded into 60 mL Bond Elute columns (Varian, Palo Alto, CA, USA). The resin was cleaned on-column using 4 M HNO_3. The resin was then rinsed with Milli-Q ultrapure water (resistivity > 18 MΩ·cm; Millipore), before being conditioned with 0.5 M NH_4OH and 2 M NH_4OH. The resin was then rinsed again with ultrapure water. After loading the sample on the column, the Cl was eluted using 3 M HNO_3.

The samples were then processed using the method described by Tosaki et al. (2011a). All samples were analyzed for $^{36}Cl/Cl$ (atomic ratio of ^{36}Cl to stable Cl) using the AMS system installed at the Tandem Accelerator Complex, University of Tsukuba (Sasa et al., 2010), along with diluted NIST ^{36}Cl standards ($^{36}Cl/Cl = 1.60 \times 10^{-12}$; Sharma et al., 1990). The overall precision of the system was better than 2% and the measurement background level was ~1×10^{-15} (Sasa et al., 2010). The calculated $^{36}Cl/Cl$ ratio of the sample includes the statistical error

derived from the uncertainties (1σ) associated with the measurement of the sample, the standard, and the blank.

3. Results and discussion

Table 1 gives the chemical composition and ^{36}Cl data of monthly precipitation samples collected over the entire sampling period from 2004 to 2009. The sample for December 2005 is omitted from Table 1 because of the extremely small amount of precipitation during this month (1.0 mm; meteorological data at the Aerological Observatory). From the measured data, the ^{36}Cl deposition fluxes were calculated according to the following equation:

$$F = \frac{C \times V}{A \times D \times 24 \times 60 \times 60} \quad (1)$$

where F is the ^{36}Cl deposition flux (atoms m^{-2} s^{-1}), C is the ^{36}Cl concentration (atoms L^{-1}), V is the collected volume of precipitation (L), A is the cross-sectional area of the top of the funnel (m^2), and D is the length of the sampling period (days). The Cl fluxes were similarly calculated from the Cl^- concentrations using Eq. 1.

To ensure sampling efficiency, the collected amount of monthly precipitation was compared with the observed meteorological data from the Aerological Observatory (Fig. 2). A portion of the sample with the greatest monthly precipitation (October 2004) was lost because of overflow from the collector during a typhoon event. For the August 2007 sample, the precipitation amount calculated from the collected volume (60.5 mm) was much greater than the observed meteorological data value (21.0 mm). These two values differ because of the spatial heterogeneity of precipitation; the observed data at ~1 km northwest of the study site was also higher than the observed meteorological data value (44.0 mm) from the Terrestrial Environment

Research Center (TERC), University of Tsukuba. The majority of the samples plot close to the line of agreement (Fig. 2), suggesting that the sampling efficiency was ~100%. Small sample losses from the collectors due to evaporation would not influence the ^{36}Cl and Cl fluxes because these fluxes are calculated from the collected precipitation volume.

The source of Cl^- in precipitation was evaluated by plotting the concentrations of elements predominately of seawater origin (Na^+ and Mg^{2+}) against Cl^- concentrations (Fig. 3). Figure 3a shows an overall enrichment in Mg^{2+} relative to seawater, which may be attributed to Mg^{2+} contributions from wind-blown soil particles. This interpretation is supported by the Ca^{2+}/Cl^- ratios, which are much higher than those of seawater (calculated from Table 1). The contribution of soil particles seems to be less dominant in high-precipitation months (Fig. 3; see the next paragraph for P/P_{avg}), probably because higher rainfall leads to less incorporation of dust. In contrast, the Na^+/Cl^- ratios of the precipitation are similar to that of seawater (Fig. 3b), which suggests that the vast majority of the Cl in the precipitation originates from seawater (sea spray). The $^{36}Cl/Cl$ ratio of seawater is close to the detection limit of AMS, 1×10^{-15} (Argento et al., 2010); consequently, the ^{36}Cl contained in the precipitation samples should reflect production in the atmosphere.

Figure 4 shows $^{36}Cl/Cl$ ratios in monthly precipitation samples plotted against $1/Cl^-$. The presence of a positive correlation confirms that $^{36}Cl/Cl$ ratios in precipitation are determined primarily by the amount of stable Cl deposition. To examine the effect of varying precipitation amounts (Fig. 4), the data were divided into two groups based on P/P_{avg}, the deviation of the monthly precipitation amount (P) from the average observed monthly precipitation value (P_{avg}) at the nearest weather station (i.e., the Aerological Observatory). The data were divided at $P/P_{avg} = 1.5$, which corresponds to a 50% increase of monthly precipitation. The samples with $P > 1.5P_{avg}$ plot below the trend formed by the samples with $P < 1.5P_{avg}$, which suggests a

precipitation dilution effect on Cl^- concentration (Fig. 4).

To exemplify the temporal variability of ^{36}Cl and Cl in precipitation, the measured data and calculated flux values for the entire sampling period are presented in Fig. 5. The $^{36}Cl/Cl$ ratio exhibits an annual peak, which occurs around June (Fig. 5d). On the other hand, the ^{36}Cl flux exhibits a peak in April–May (Fig. 5f). This difference is attributed to the significant decrease of Cl flux in June (Fig. 5c), which leads to an increase in the $^{36}Cl/Cl$ ratio. The most probable cause of this phenomenon is the washout effect by the persistent rain during the East Asian rainy season, which generally occurs from June to mid-July in central Japan. The Cl and ^{36}Cl concentrations were not characterized by obvious seasonal trends (Fig. 5b and e, respectively) because the precipitation dilution effect (discussed above) directly affects both concentrations (Fig. 4).

Similar spring maxima in the ^{36}Cl flux have been observed at other locations (Hainsworth et al., 1994; Knies et al., 1994) and for other cosmogenic radionuclides; e.g., ^{7}Be (e.g., Viezee and Singh, 1980) and ^{10}Be (e.g., Raisbeck et al., 1979). As discussed in these previous studies, the seasonal pattern is attributed to annual variation in the tropopause height (Staley, 1962). Since the tropopause begins to rise rapidly in April or May at mid-latitudes (especially at 30–40°N), stratospheric air masses, rich in ^{36}Cl, are transported into the troposphere and washed out of the atmosphere after approximately 1 week. Figure 6 shows the average monthly values (calculated from the measured data) and corroborates the existence of seasonal trends as described above. Observed increases in the ^{36}Cl flux, other than those that occur in the spring (e.g., October 2004, August 2005, and August 2008; Fig. 5f), are attributed to precipitation amounts (Fig. 5a). This interpretation is supported by the empirical dependence of atmospheric fallout of cosmogenic nuclides on precipitation amount (Knies et al., 1994; Phillips, 2000). However, the correlation between precipitation amount and ^{36}Cl flux is weak, as it is masked by seasonal variation.

The seasonal variation is likely to cause a pronounced $^{36}Cl/Cl$ peak signal in groundwater recharge between May and June, suggesting that several years would be required to constrain the local initial $^{36}Cl/Cl$ value and the annual average ^{36}Cl flux. The measured monthly ^{36}Cl fluxes were averaged for each year to obtain a precise estimate of annual average ^{36}Cl flux (Table 2). The flux values were relatively stable between 2004 and 2007, while they increased for 2008 and 2009 (Fig. 7). This increase may be related to variation in solar activity, as observed for ^{7}Be (Cannizzaro et al., 2004; Kulan et al., 2006). The smallest annual sunspot numbers were observed for 2008 and 2009 (Fig. 7), which leads to an increase in the production of nuclides due to increased galactic cosmic ray fluxes, while ^{36}Cl production by solar proton is negligible (Huggle et al., 1996). As this period may be regarded as an extreme event, the mean value for 2004–2007 (32 ± 2 atoms m^{-2} s^{-1}) is expected to be closer to the long-term average flux. This value is in good agreement with the latitudinal dependence of ^{36}Cl fallout (Lal and Peters, 1967), after normalization to the global mean production rate of 20 atoms m^{-2} s^{-1} (cf. Huggle et al., 1996; Masarik and Beer, 1999, 2009). Therefore, we conclude that chlorine recycling via CH_3Cl is not evident at present in central Japan.

Finally, the annual $^{36}Cl/Cl$ values were averaged for 2004–2007 to obtain an estimate of the initial $^{36}Cl/Cl$ value, $(41 \pm 6) \times 10^{-15}$. This value is comparable to that of pre-bomb groundwater sampled from an observation well on the premises of the Geological Survey of Japan (sampling depth ~50 m) at the Tsukuba Upland, $(31 \pm 3) \times 10^{-15}$. However, Table 2 shows that an even longer period, covering an 11-year solar cycle, is required to obtain a well-constrained initial value. For further consideration of the relationship between the ^{36}Cl flux and solar activity, continuous precipitation sampling is ongoing by the authors.

4. Conclusions

The ^{36}Cl deposition flux in central Japan was continuously observed for >5 years. The chlorine in the precipitation was identified as originating from sea spray, indicating that ^{36}Cl produced in the atmosphere dominates in the precipitation in the study area. The ^{36}Cl flux showed a clear seasonal variation with an annual peak during the spring, which is attributed to the seasonal variability of tropopause height. The long-term average ^{36}Cl flux estimated from the measured data is consistent with the prediction from the ^{36}Cl latitudinal fallout model scaled using the global mean production rate of 20 atoms m^{-2} s^{-1}. The initial $^{36}Cl/Cl$ ratio was estimated to be $(41 \pm 6) \times 10^{-15}$, which is similar to that of pre-bomb groundwater in the Tsukuba Upland. The increase in the ^{36}Cl flux between 2008 and 2009 may have resulted from decreased solar activity during that time. We will further investigate this relationship by continuing the precipitation monitoring for several more years.

Acknowledgements

We are grateful to Dr. Kunihiko Nishiizumi for his advice on sample preparation using ion exchange chromatography. Dr. Riki Seki is gratefully acknowledged for her support in the development of the ion exchange system. Ms. Satomi Abe is acknowledged for her help with the preparation of precipitation samples for AMS. The precipitation collector used in this study was designed based on advice from Dr. Shiho Yabusaki. We thank all the members of the AMS Group at the University of Tsukuba for their assistance with the ^{36}Cl measurements. We also thank all the staff at the Tandem Accelerator Complex (University of Tsukuba) for their support during accelerator operation. This research was supported in part by a research grant held by K.S. from the Japan Science and Technology Agency (JST), Grants-in-Aid for Scientific Research (B) (Nos. 21310004, 19300304, and 18360043) from the Japan Society for the Promotion of Science (JSPS), and a research grant (No. 22343) from the Kurita Water and

Environment Foundation (KWEF), Japan.

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5

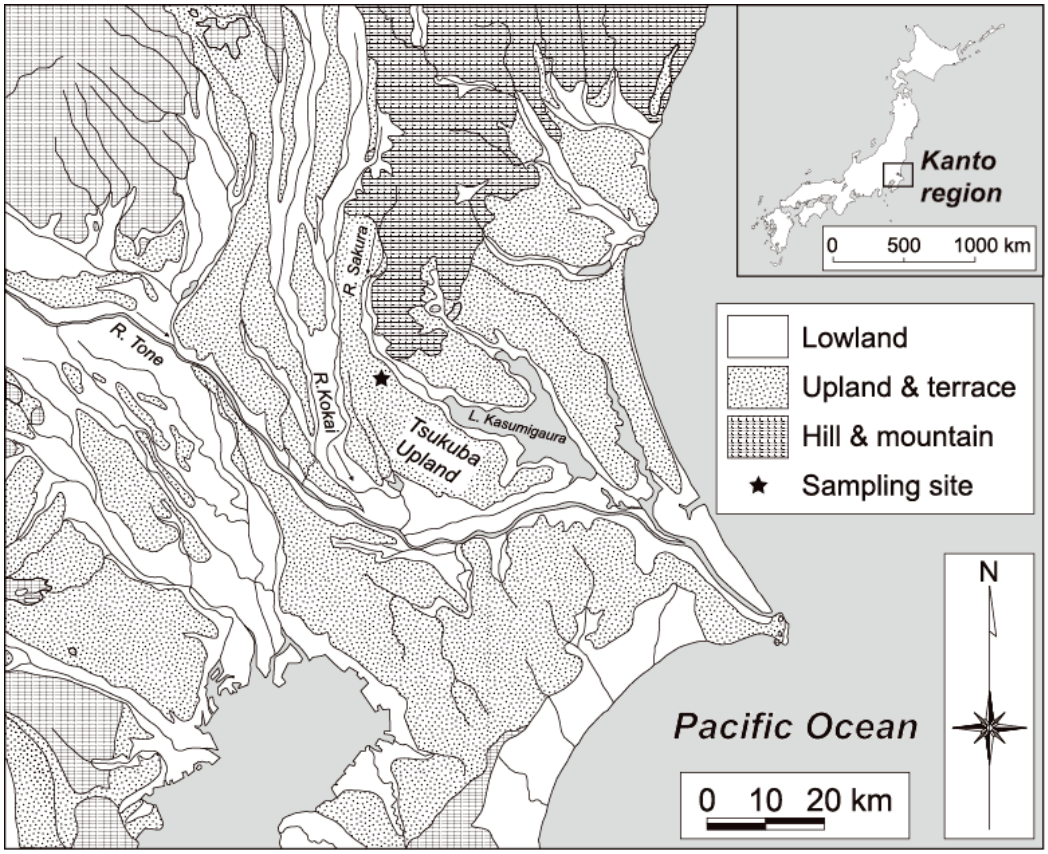

Figure 1. Location of the precipitation sampling site.

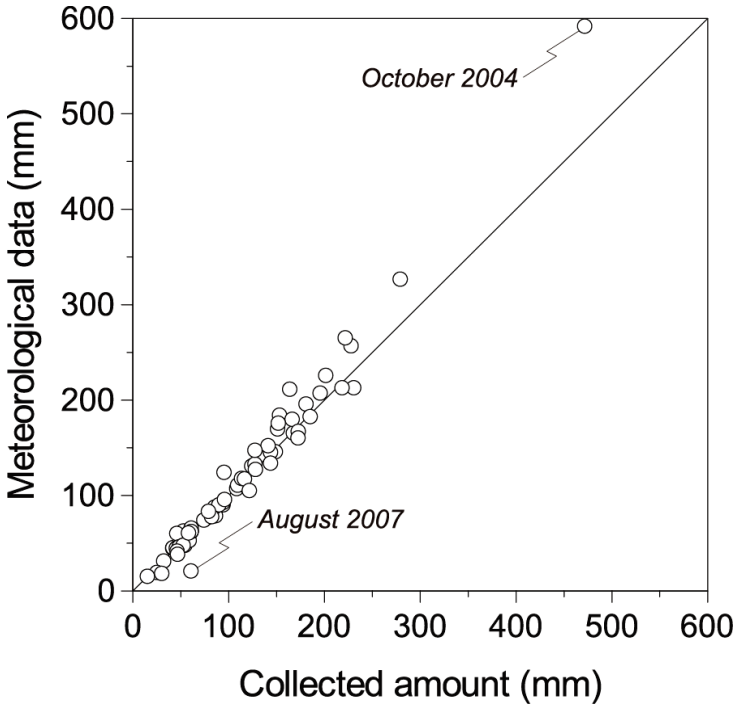

Fig. 2. Comparison between the measured precipitation amount and meteorological data.

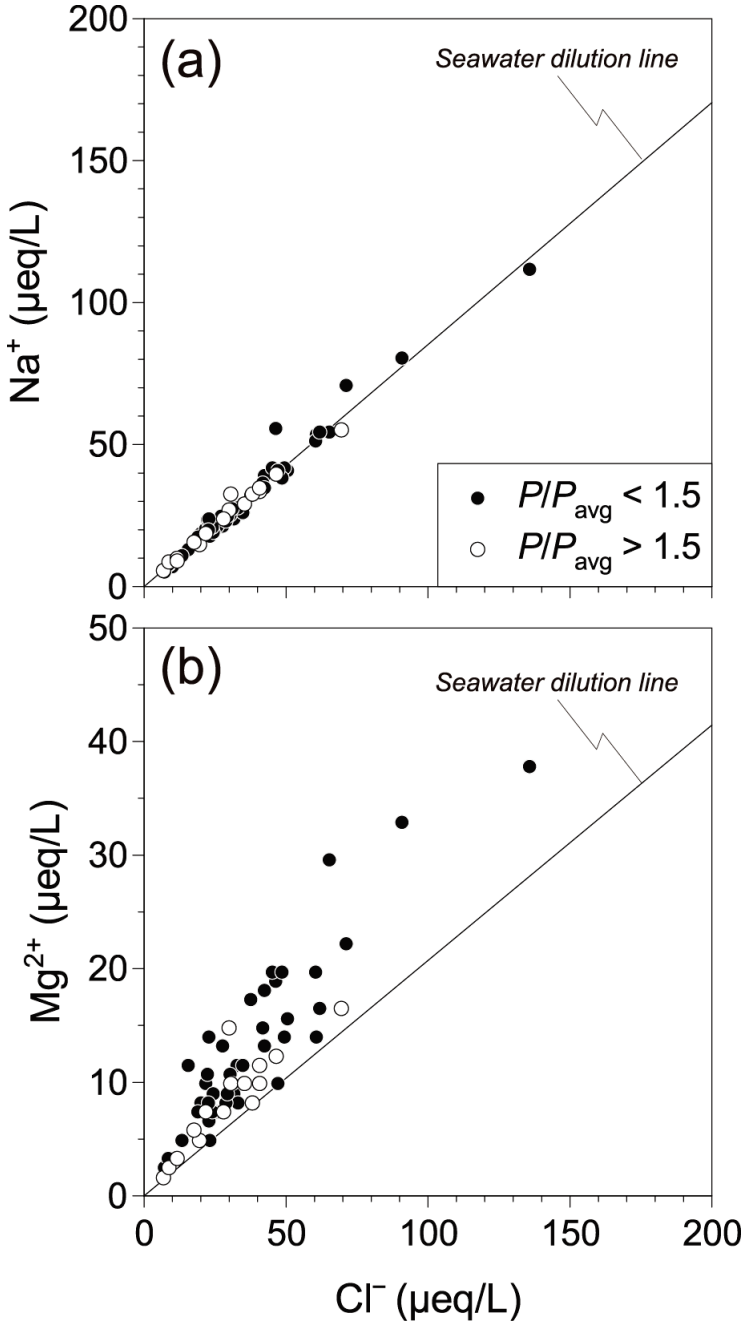

Fig. 3. (a) Na^+ concentrations and (b) Mg^{2+} concentrations plotted against Cl^- concentrations in precipitation. P/P_{avg} denotes the deviation of the monthly precipitation amount (P) from the average observed monthly precipitation value (P_{avg}) at the nearest weather station (see the text for details).

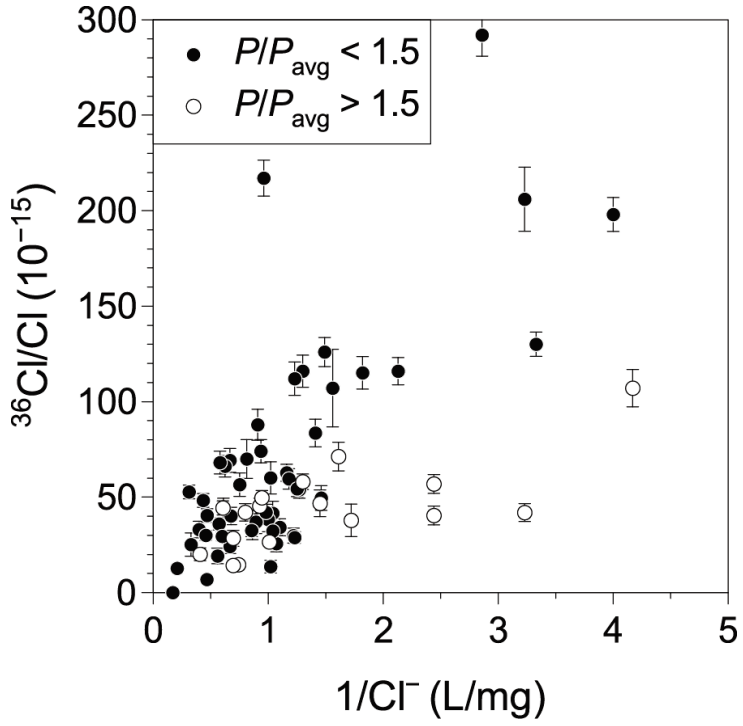

Fig. 4. ^{36}Cl/Cl ratios in precipitation plotted against the reciprocal of Cl^- concentrations ($1/Cl^-$). P/P_{avg} denotes the deviation of the monthly precipitation amount (P) from the average observed monthly precipitation value (P_{avg}) at the nearest weather station. The error bars represent the errors involved in measuring ^{36}Cl by accelerator mass spectrometry (see the text for details).

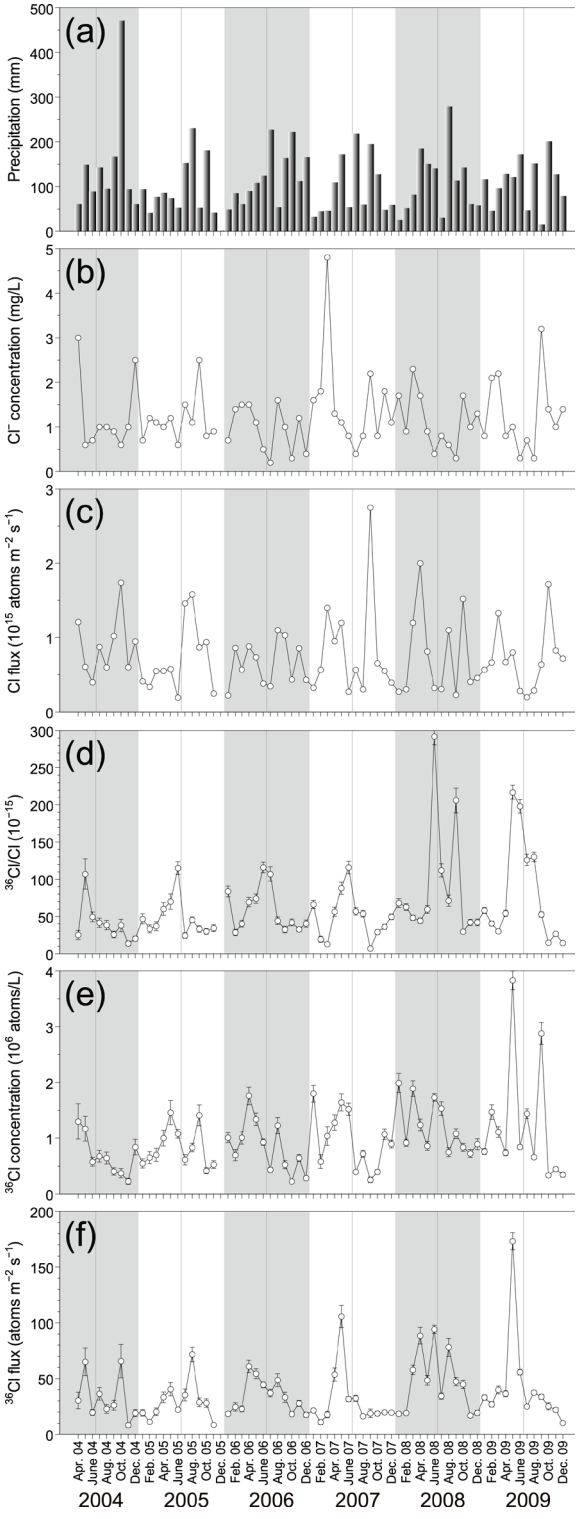

Fig. 5. Temporal variations in (a) precipitation amount, (b) Cl^- concentration, (c) Cl flux, (d) $^{36}Cl/Cl$ ratio, (e) ^{36}Cl concentration, and (f) ^{36}Cl flux. Shading and vertical lines were added to visually separate each observation year.

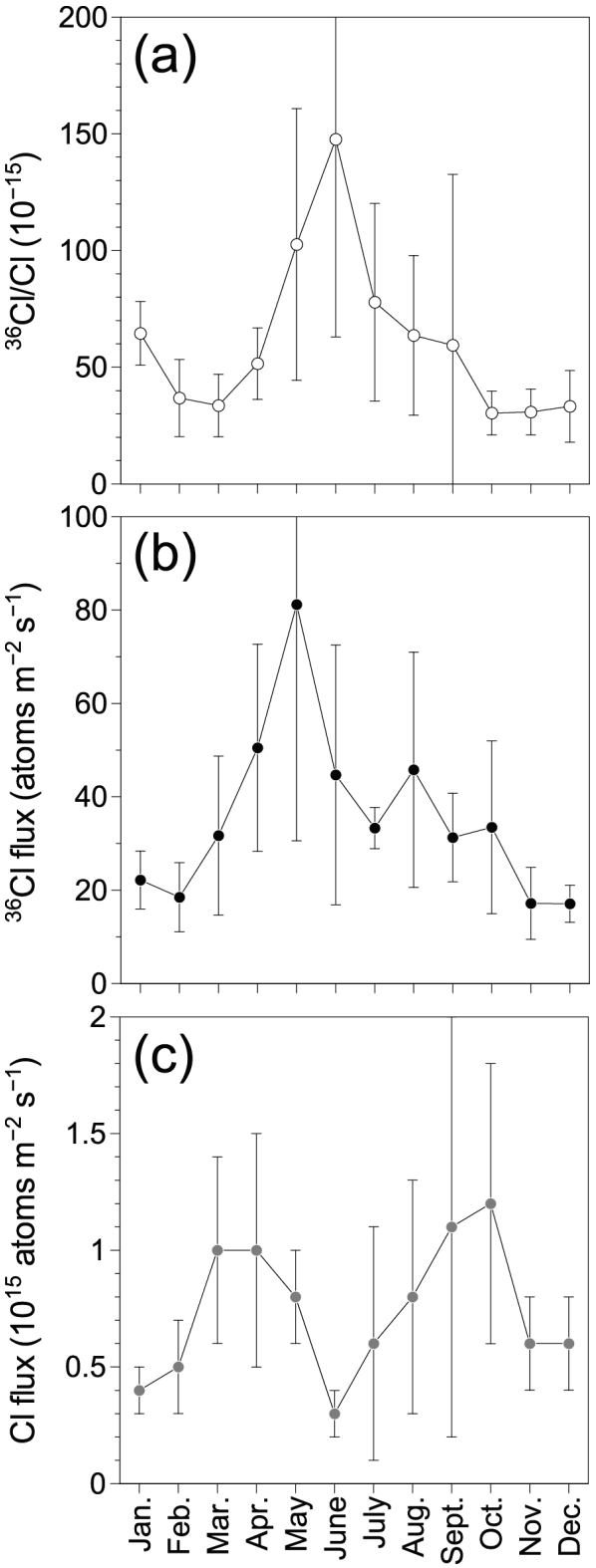

Fig. 6. Seasonal variations in (a) ^{36}Cl/Cl ratio, (b) ^{36}Cl flux, and (c) Cl flux. The error bars indicate the standard deviation of 5 or 6 values for each month.

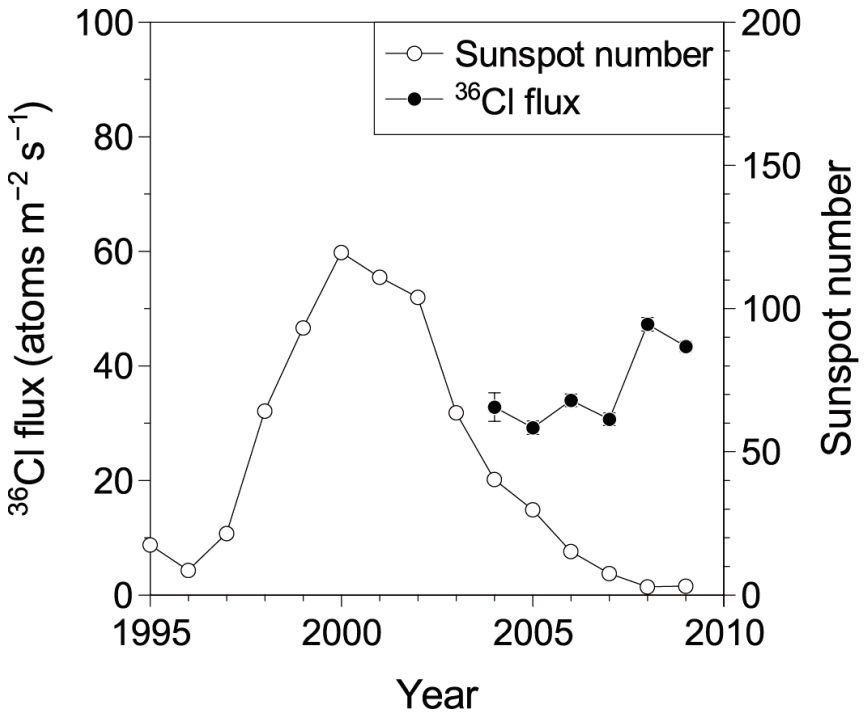

Fig. 7. Comparison between the annual average ^{36}Cl flux and the sunspot number (data from the Solar Influences Data Analysis Center, Belgium).

1 Table 1. Chemical composition and ^{36}Cl data for monthly precipitation samples at Tsukuba.

Month	Precipitation (mm)	Na^+ (mg L^{-1})	Mg^{2+} (mg L^{-1})	Ca^{2+} (mg L^{-1})	Cl^- (mg L^{-1})	Cl flux (10^{15} atoms m^{-2} s^{-1})	$^{36}Cl/Cl$ ratio (10^{-15})	^{36}Cl concentration (10^6 atoms L^{-1})	^{36}Cl flux (atoms m^{-2} s^{-1})
Apr. 04	60.5	–	–	–	3.04	1.21	25 ± 6	1.30 ± 0.31	30.3 ± 7.4
May 04	148.7	–	–	–	0.64	0.61	107 ± 20	1.17 ± 0.22	65.0 ± 12.3
June 04	88.6	–	–	–	0.69	0.40	50 ± 6	0.58 ± 0.08	19.7 ± 2.6
July 04	143.5	–	–	–	0.96	0.87	42 ± 6	0.68 ± 0.10	36.3 ± 5.6
Aug. 04	95.0	–	–	–	1.00	0.60	38 ± 6	0.65 ± 0.11	22.9 ± 3.8
Sept. 04	167.5	–	–	–	0.93	1.02	26 ± 4	0.40 ± 0.07	26.2 ± 4.4
Oct. 04	471.4	–	–	–	0.58	1.74	38 ± 9	0.37 ± 0.08	65.6 ± 14.8
Nov. 04	94.0	–	–	–	0.98	0.60	14 ± 3	0.23 ± 0.06	8.2 ± 2.0
Dec. 04	60.8	1.27	0.20	0.62	2.46	0.95	20 ± 3	0.84 ± 0.14	19.1 ± 3.1
Jan. 05	94.1	0.34	0.06	0.32	0.69	0.41	47 ± 7	0.55 ± 0.08	19.3 ± 2.9
Feb. 05	41.4	0.58	0.14	0.82	1.16	0.34	33 ± 5	0.65 ± 0.11	11.2 ± 1.8
Mar. 05	77.4	0.55	0.11	0.56	1.12	0.55	37 ± 6	0.70 ± 0.12	20.3 ± 3.4

Apr. 05	86.0	0.49	0.16	0.99	0.98	0.55	60 ± 8	1.00 ± 0.14	33.2 ± 4.7
May 05	73.9	0.60	0.14	0.71	1.23	0.58	70 ± 10	1.46 ± 0.21	40.4 ± 5.9
June 05	53.4	0.30	0.14	1.09	0.55	0.19	115 ± 8	1.08 ± 0.08	22.2 ± 1.6
July 05	153.0	0.90	0.16	0.34	1.50	1.46	24 ± 4	0.62 ± 0.09	35.2 ± 5.3
Aug. 05	230.5	0.75	0.12	0.26	1.08	1.58	45 ± 4	0.83 ± 0.07	71.6 ± 6.4
Sept. 05	52.5	1.63	0.27	0.78	2.52	0.87	33 ± 4	1.41 ± 0.19	28.7 ± 3.8
Oct. 05	180.5	0.41	0.06	0.14	0.82	0.94	30 ± 4	0.42 ± 0.06	28.1 ± 3.7
Nov. 05	41.6	0.49	0.09	0.31	0.91	0.25	34 ± 4	0.53 ± 0.07	8.5 ± 1.1
Dec. 05	–	–	–	–	–	–	–	–	–
Jan. 06	49.2	0.43	0.10	0.82	0.71	0.22	84 ± 7	1.01 ± 0.09	18.5 ± 1.6
Feb. 06	85.2	0.77	0.12	0.43	1.44	0.86	29 ± 4	0.70 ± 0.10	24.6 ± 3.6
Mar. 06	60.8	0.84	0.18	1.19	1.48	0.57	40 ± 5	1.01 ± 0.11	22.9 ± 2.6
Apr. 06	89.7	0.80	0.22	1.21	1.50	0.88	69 ± 6	1.76 ± 0.16	61.0 ± 5.5
May 06	108.3	0.58	0.13	0.58	1.07	0.73	74 ± 6	1.34 ± 0.11	54.4 ± 4.5
June 06	123.9	0.25	0.06	0.31	0.47	0.38	116 ± 7	0.93 ± 0.06	44.4 ± 2.7

July 06	227.5	0.13	0.02	0.13	0.24	0.35	107 ± 10	0.44 ± 0.04	37.0 ± 3.4
Aug. 06	54.0	1.28	0.23	0.64	1.64	1.10	44 ± 5	1.23 ± 0.15	48.7 ± 5.8
Sept. 06	163.6	0.57	0.09	0.17	0.96	1.03	32 ± 4	0.53 ± 0.07	33.2 ± 4.5
Oct. 06	221.5	0.20	0.03	0.10	0.31	0.44	42 ± 5	0.22 ± 0.02	18.3 ± 2.0
Nov. 06	111.6	0.64	0.10	0.27	1.17	0.86	32 ± 3	0.65 ± 0.06	27.8 ± 2.7
Dec. 06	166.2	0.23	0.04	0.20	0.41	0.43	40 ± 5	0.28 ± 0.03	17.5 ± 2.1
Jan. 07	32.0	0.96	0.24	1.40	1.60	0.33	66 ± 6	1.80 ± 0.15	21.5 ± 1.8
Feb. 07	45.1	0.94	0.19	0.84	1.79	0.57	19 ± 4	0.58 ± 0.12	10.9 ± 2.3
Mar. 07	45.8	2.57	0.46	1.81	4.81	1.40	13 ± 2	1.04 ± 0.17	17.8 ± 2.9
Apr. 07	109.1	0.74	0.21	1.26	1.33	0.95	56 ± 6	1.28 ± 0.14	53.7 ± 5.8
May 07	172.4	0.63	0.12	0.46	1.10	1.20	88 ± 8	1.64 ± 0.15	105.7 ± 9.8
June 07	54.0	0.47	0.12	0.91	0.77	0.27	116 ± 8	1.52 ± 0.11	31.6 ± 2.3
July 07	218.1	0.21	0.04	0.16	0.41	0.57	57 ± 5	0.40 ± 0.03	32.3 ± 2.8
Aug. 07	60.5	0.54	0.13	0.94	0.79	0.30	54 ± 4	0.72 ± 0.06	16.3 ± 1.3
Sept. 07	195.2	1.23	0.17	0.48	2.15	2.75	7 ± 1	0.25 ± 0.05	18.9 ± 3.8

Oct. 07	127.3	0.52	0.08	0.18	0.81	0.65	29 ± 3	0.40 ± 0.04	18.8 ± 2.0
Nov. 07	48.2	0.96	0.17	0.56	1.75	0.55	36 ± 3	1.07 ± 0.09	19.8 ± 1.8
Dec. 07	58.6	0.62	0.18	0.80	1.06	0.39	50 ± 4	0.89 ± 0.07	19.6 ± 1.5
Jan. 08	24.9	0.88	0.24	1.25	1.72	0.27	68 ± 6	1.99 ± 0.17	18.5 ± 1.6
Feb. 08	52.3	0.44	0.11	0.66	0.86	0.30	63 ± 4	0.92 ± 0.06	19.1 ± 1.3
Mar. 08	82.0	1.25	0.36	1.64	2.31	1.20	48 ± 4	1.89 ± 0.14	57.9 ± 4.2
Apr. 08	184.9	0.91	0.15	0.34	1.65	2.00	44 ± 4	1.24 ± 0.10	88.5 ± 7.5
May 08	150.9	0.48	0.09	0.39	0.85	0.81	60 ± 5	0.86 ± 0.08	48.5 ± 4.4
June 08	140.9	0.16	0.04	0.25	0.35	0.32	292 ± 11	1.73 ± 0.07	94.2 ± 3.6
July 08	30.0	0.55	0.17	1.08	0.81	0.31	112 ± 9	1.53 ± 0.12	34.3 ± 2.7
Aug. 08	278.9	0.36	0.07	0.19	0.62	1.10	71 ± 8	0.75 ± 0.08	78.1 ± 8.2
Sept. 08	113.2	0.18	0.04	0.24	0.31	0.23	206 ± 17	1.08 ± 0.09	47.3 ± 3.9
Oct. 08	143.5	0.94	0.12	0.22	1.67	1.52	29 ± 2	0.84 ± 0.07	44.8 ± 3.7
Nov. 08	60.6	0.53	0.10	0.40	1.02	0.41	42 ± 4	0.73 ± 0.07	17.0 ± 1.7
Dec. 08	57.9	0.67	0.12	0.61	1.25	0.46	42 ± 5	0.89 ± 0.10	19.3 ± 2.1

Jan. 09	116.4	0.43	0.09	0.42	0.77	0.57	58 ± 4	0.76 ± 0.05	33.0 ± 2.3
Feb. 09	45.9	1.18	0.24	1.27	2.14	0.67	40 ± 4	1.47 ± 0.13	26.9 ± 2.4
Mar. 09	95.6	1.25	0.20	0.69	2.19	1.33	30 ± 3	1.11 ± 0.10	39.8 ± 3.5
Apr. 09	127.8	0.46	0.10	0.67	0.80	0.67	54 ± 4	0.74 ± 0.05	36.4 ± 2.7
May 09	121.3	0.57	0.11	0.46	1.04	0.80	217 ± 9	3.83 ± 0.17	173.2 ± 7.5
June 09	172.1	0.12	0.03	0.19	0.25	0.28	198 ± 9	0.84 ± 0.04	55.8 ± 2.5
July 09	46.6	0.40	0.09	0.73	0.67	0.20	126 ± 8	1.44 ± 0.09	25.0 ± 1.5
Aug. 09	151.6	0.18	0.04	0.26	0.30	0.29	130 ± 6	0.66 ± 0.03	37.4 ± 1.8
Sept. 09	15.1	1.85	0.40	2.16	3.22	0.64	53 ± 4	2.88 ± 0.19	33.6 ± 2.2
Oct. 09	201.2	0.75	0.10	0.17	1.35	1.72	15 ± 2	0.34 ± 0.04	25.2 ± 3.1
Nov. 09	127.3	0.55	0.09	0.26	0.99	0.83	27 ± 2	0.45 ± 0.04	22.0 ± 1.9
Dec. 09	78.7	0.80	0.14	0.27	1.44	0.72	14 ± 2	0.35 ± 0.05	10.2 ± 1.3

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2

1 Table 2. Annual average ^{36}Cl flux, Cl flux, and $^{36}Cl/Cl$ ratio.

Year	^{36}Cl flux (atoms m^{-2} s^{-1})	Cl flux (10^{15} atoms m^{-2} s^{-1})	$^{36}Cl/Cl$ (10^{-15})
2004[a]	33 ± 3	0.89	37 ± 3
2005[b]	29 ± 1	0.71	41 ± 2
2006	34 ± 1	0.65	52 ± 2
2007	31 ± 1	0.83	37 ± 1
2008	47 ± 1	0.75	63 ± 2
2009	43 ± 1	0.73	60 ± 1
Average for 2004–2007	32 ± 2	0.77 ± 0.11	41 ± 6
Average for 2004–2009	36 ± 7	0.76 ± 0.09	48 ± 11

[a]Average for April–December 2004.

[b]Average for January–November 2005.

The error on the average values is reported as the standard deviation of the annual values.

2